Diffusion Studies with X-Ray Photon Correlation Spectroscopy at Present and Future Light Sources

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The dynamical properties of hard and soft condensed matter are of considerable importance from a fundamental point of view, but are in addition essential for the technical applications of bulk or nanoscale materials. Macroscopically, diffusion can be mathematically described (Fick's laws) and investigated with standard methods like tracer diffusion experiments, for a review, see Mehrer [1]. However, the underlying processes on an atomic level, i.e. atomic jump vectors and frequencies, are not easily accessible. Up to now only quasi-elastic methods, particularly quasi-elastic neutron scattering (QNS), quasi-elastic Mößbauer spectroscopy (QMS), nuclear resonant scattering (NRS) of synchrotron radiation, and (most recently) the neutron-spin echo technique, were capable of resolving the elementary atomic jump process (for a review, see Vogl and Sepiol [1]). The above mentioned techniques suffer, however, from two big disadvantages. First, only certain isotopes are "visible" for each technique, e.g. the Mößbauer isotope ^{57}Fe in case of QMS and NRS. Second, due to a limited energy resolution of all techniques only very fast diffusion is detectable by the above-mentioned methods. Hence, a non-resonant technique – not restricted to certain isotopes–, which can detect slow diffusion, is extremely desirable. The emerging method of X-ray photon correlation spectroscopy (XPCS) [2] is the most promising candidate to fill this gap. Basically the accessible wavelength and coherency of modern 3rd generation synchrotron sources should be sufficient to resolve the jump process on the atomic level, however, as the coherent intensity in the hard X-ray regime is limited, one has not succeeded until now in experimental verification. Having the forthcoming extremely brilliant fourth-generation sources at the horizon, a serious assessment of possible diffusion studies seems to be mandatory. This next generation of sources, based on a free-electron laser, is already operating and will culminate with the hard X-ray FEL's expected to become operational near the end of this decade.
Here we present a theoretical treatment of the expected signal-to-noise ratio given a certain count rate, that is we deliver estimates on the minimum count rates for measuring atomic diffusion and present recipes for optimising the signal-to-noise ratio. Also we examine samples concerning their expected scattered intensity and the possibility of resolving atomic diffusion at today's synchrotrons. Furthermore, an outlook is given on the possibility of such measurements at the European X-ray free electron laser [3].

[1] P. Heitjans and J. Kärger, (Eds.), *Diffusion in Condensed Matter* (Springer, Berlin/Heidelberg, 2005).
[2] G. Grübel and F. Zontone, J. Alloy. Compd. **362**, 3 (2004).
[3] *XFEL Technical Design Report* (2006): http://xfel.desy.de/tdr.